

# Sign reversal of the Hall resistance in the mixed-state of $\text{La}_{1.89}\text{Ce}_{0.11}\text{CuO}_4$ and $\text{La}_{1.89}\text{Ce}_{0.11}(\text{Cu}_{0.99}\text{Co}_{0.01})\text{O}_4$ thin films

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## Abstract

The transport properties of  $\text{La}_{1.89}\text{Ce}_{0.11}\text{CuO}_4$  (LCCO) and  $\text{La}_{1.89}\text{Ce}_{0.11}(\text{Cu}_{0.99}\text{Co}_{0.01})\text{O}_4$  (LCCO:Co) superconducting thin films are investigated. When the external field  $\mathbf{H}$  is applied along the crystallographic  $c$ -axis, a double sign reversal of the Hall voltage in the mixed state of LCCO:Co thin films is observed whereas a single sign reversal is detected in LCCO. A double sign reversal of the Hall signal in LCCO can be recovered if the magnetic field is tilted away from the plane of the film. We find that the transition from one to two of the Hall sign reversal coincides with the change in the pinning from strong to weak. This temperature/field induced transition is caused either by the magnetic impurities in LCCO:Co or by the coupling between the pancake vortices and the in-plane Josephson vortices in LCCO. These results are in agreement with early theoretical and numerical predictions.

*Key words:* Flux pinning, magnetic properties, cuprate superconductors, superconducting films

*PACS:* 74.25.Qt, 74.25.Ha, 74.72.-h, 74.78.-w

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## 1. Introduction

Hall effect is considered as a powerful method to probe the Fermi surface of metallic compounds in general, and particularly useful to identify the nature of the charge carriers in non-magnetic systems[1–5]. However, in the mixed state of superconductors, the Hall voltage is mainly determined by the vortex motion along the direction of the bias current flow[6]. In these systems, a long standing unsolved issue is the presence of a sign reversal in the Hall voltage when changing either temperature or magnetic field.

There have been many experiments and theoretical studies focused on this so called anomalous Hall effect. For example, sign reversals has been reported in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  (YBCO) crystals [7,8]

and  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  thin films [9], and double sign reversal and even triple sign reversal have been observed in  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$  (TBCCO) [10] and  $\text{HgBa}_2\text{CaCu}_2\text{O}_6$  thin films [11], respectively. Several models, based on two-band [12], thermal fluctuation [13], pinning effect [6,14], and vortex interaction [15,16], have been proposed to interpret the Hall anomalies. In electron-doped High- $T_c$  superconductors, a distinct feature revealed by the angular-resolution photoemission experiments [17] is the coexistence of the hole- and electron-bands near the optimal doping region, which may account for the sign reversal of the Hall resistivity for temperatures above the superconducting transition  $T_c$  [18]. However, for explaining the presence of sign reversals below  $T_c$ , it is still necessary to discuss them in terms of vortex pinning mechanisms which

have been demonstrated to play a key role [14,15].

Indeed, in a seminal work, Wang and Ting [19] demonstrated that the Hall resistivity  $\rho_{xy}$  as a function of the magnetic field in the flux flow regime exhibits a sign change due to backflow currents arising from pinning forces. However, Vinokur, Geshkenbein, Feigel'man, and Blatter [6] on the basis of a phenomenological model which included pinning and thermal fluctuation, concluded that the magnitude of the Hall angle  $\Theta_H$ , does not depend on the pinning. Whether pinning is necessary for the anomalous Hall effect or not is still a fundamental question, although there is clear experimental evidence indicating that the abnormal Hall effect is indeed pinning dependent [7].

Based on their former work, Wang, Dong, and Ting (WDT) developed a unified theory for the flux motion [14] by taking into account vortex pinning and thermal fluctuations which could explain both the single and the double sign reversals of the Hall resistivity due to different pinning strength. The WDT prediction has been further confirmed by Zhu *et al.* by a numerical simulation [20]. Their work has clearly shown that the double sign change of  $\rho_{xy}$  versus the temperature appears in the weak pinning environment, while the single sign reversal occurs in the strong pinning system. Importantly, the authors have also found that there are two distinctive weak pinning configurations which can achieve the double sign reversal of  $\rho_{xy}(T)$ , i.e., one possesses low density of the pinning centers but with strong individual pinning potentials, whereas the other consists of a high pinning concentration of weak individual pinning potentials. An elegant experimental study by Kang *et al.* [7] unambiguously showed the relevance of the pinning strength for the sign reversal of the Hall resistance by irradiating a sample with heavy-ion to progressively increase the pinning strength. What it is still lacking is a more tunable pinning which allows one to go from the strong limit to the weak limit. In this case, one may expect a transition from the single sign reversal to the double sign reversal in the Hall resistivity  $\rho_{xy}$ .

In the present work, we present experimental evidence on the relevance of the pinning strength in determining the number of sign reversals in the Hall voltage. We show that despite the fact that LCCO thin films exhibit only one sign reversal with the applied field perpendicular to the *ab*-plane, a second sign reversal emerges either when rotating the magnetic field and maintaining the same perpendicular component or by changing the pinning

strength by substituting Co for Cu in order to obtain  $\text{La}_{1.89}\text{Ce}_{0.11}(\text{Cu}_{0.99}\text{Co}_{0.01})\text{O}_4$  (LCCO:Co). These two different methods show that the transition of the Hall sign reversals from single to double, coincides with the change in the pinning environment from strong to weak. Our experimental results are in agreement with previous prediction based on analytical and numerical investigations [14,20].

## 2. Experimental details

All the LCCO thin films used in the present work were fabricated by a dc magnetron sputtering method [21,22]. The transport measurements of both LCCO and LCCO:Co thin films were carried out by using a commercial Quantum Design PPMS-14. More details about the film preparation and the measurement setup can be found elsewhere [18,21–26]. All the thin films were patterned by photolithography and subsequent ion milling, into bridges of 2100  $\mu\text{m}$  long, 100  $\mu\text{m}$  wide and with six terminals. The critical transition temperatures  $T_{c0}$  for LCCO and LCCO:Co are  $\sim 24$  and 13 K as reported in our previous work [23], respectively. The upper critical magnetic field  $H_{c2}(0)$  for LCCO is about 10 Tesla [18].

## 3. Results and discussions

Let us start by demonstrating experimentally that the tilting of the magnetic field with respect to the plane of a thin LCCO film leads to significant changes in the pinning properties as well as the sign reversal of the Hall resistivity  $\rho_{xy}$ . In order to investigate this effect we study the Hall resistivity  $\rho_{xy}$  and longitudinal resistivity  $\rho_{xx}$  as a function of the perpendicular field  $B_z$  by either varying the strength but maintaining the field orientation perpendicular to the *ab*-plane (mode A) or by changing the angle  $\theta$  between the field and the *ab*-plane with constant field intensity (mode B). During the measurements, the applied current  $j$  in the *ab*-plane is always perpendicular to the magnetic field, i.e.,  $\mathbf{j} \perp \mathbf{B}$ . This method allows us to discern the effects arising purely from the out of the plane component of the applied field from those originated by the in-plane field.

First we would like to point out that in the LCCO sample and for  $T > T_c$ , both  $\rho_{xy}(B_z)$  and  $\rho_{xx}(B_z)$  curves overlap with each other no matter whether the perpendicular component of the magnetic field

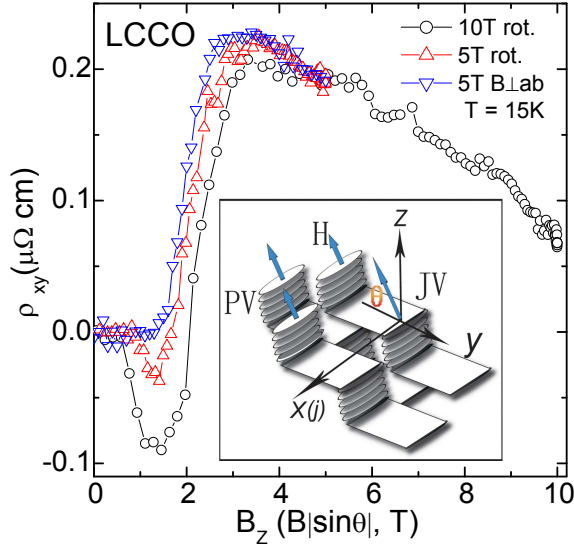


Fig. 1. (Color online) Hall resistivity  $\rho_{xy}$  versus  $B_z$  ( $B|\sin\theta|$ ) in LCCO thin films at 15 K by different field changing modes. Inset: illustration of tilted vortices with JV representing in-plane Josephson vortices and PV representing the pancake vortices.

changes its magnitude or its orientation [18]. This indicates that the contribution of parallel-field component ( $B_y$ ) to the magnetoresistance is negligible in comparison with  $B_z$ . The question naturally arises whether this is also true for the case of  $T < T_{c0}$ . In Fig. 1, isothermal curves of  $\rho_{xy}$  versus  $B_z = B|\sin\theta|$  for LCCO are plotted at 15 K ( $< T_{c0}$ ). Here, we find that  $\rho_{xy}(B_z)$  curve does not follow the scaling with  $B_z$  as well as the  $\rho_{xx}(B_z)$  curve as reported in Ref. [18]. Notice that  $\rho_{xy}(B_z)$  with  $\mathbf{B} \perp \mathbf{ab}$ -plane has the maximum value at  $\sim 3.5$  T and drops to zero when the magnetic field is decreased down to 1.4 T without reversing its sign, whereas the  $\rho_{xy}(B)$  measurement via rotation of the field exhibits a clear sign change from positive to negative in the low field region.

This result indicates the importance of the in-plane component of the field in the mixed state. When the magnetic field is tilted away from the  $c$ -axis, two different kinds of vortices appear in the samples [27–29], namely pancake vortices with the field component perpendicular to the  $ab$ -plane, and Josephson vortices with the component parallel to the  $ab$ -plane, as shown in the inset of Fig. 1. By rotating the field from in-plane to out-of-plane, the vortices change from Josephson vortex to pancake vortex type and vice versa.

In principle, the pinning effect for the in-plane vortices is always stronger than that for out-of-plane

vortices, i.e., it is harder to drive the Josephson vortices out of the  $ab$ -plane. Therefore, a hysteresis in the depinning field may appear [30]. Based on this consideration it is clear that changing the field orientation from  $\theta = -90^\circ$  to  $\theta = 90^\circ$ , we should be able to observe the difference between  $\rho_{xx,+\theta}$  and  $\rho_{xx,-\theta}$  due to the different evolution processes of the vortices. Here,  $\rho_{xx,+\theta}$  and  $\rho_{xx,-\theta}$  correspond to the resistivity values with  $\theta$  varying from  $0^\circ$  to  $90^\circ$  and from  $-90^\circ$  to  $0^\circ$ , respectively. The experimental data seem to support this idea. In Fig. 2 (a), the difference between  $\rho_{xx,+\theta}$  and  $\rho_{xx,-\theta}$ , i.e.,  $\Delta\rho_{xx} = \rho_{xx,+\theta} - \rho_{xx,-\theta}$ , is plotted as a function of  $B|\sin\theta|$  at 15 K with fixed magnetic field strength  $H = 10$  T. The maximum  $|\Delta\rho_{xx}|$  can reach up to  $0.03$   $m\Omega \cdot cm$  at  $B|\sin\theta| \sim 1.8$  T ( $\theta \sim 10^\circ$ ), which is about 18% of the normal-state resistivity. With the increase of the field component along the  $c$ -axis, the sample is gradually driven into the normal state and  $\Delta\rho_{xx}$  approaches zero. This demonstrates that the in-plane field component plays an important role in the mixed state.

In Fig. 2 (b), the curves of  $\rho_{xx}$  versus  $B|\sin\theta|$  are shown for different modes, i.e., either rotating the orientation or changing the strength of the field. It is clear that for the same  $B_z$ , the critical vortex depinning field for mode A with  $H \perp ab$  is higher than that for mode B by rotating field, where there is additional field component parallel to the  $ab$ -plane. The most important feature here is that the critical vortex depinning field for  $\rho_{xx,+\theta}$  from  $0^\circ$  to  $90^\circ$  is obviously larger than that for  $\rho_{xx,-\theta}$  from  $-90^\circ$  to  $0^\circ$ .

It is important to mention that the  $\rho_{xx}$  curve obtained via mode A for  $H \perp ab$  overlaps with the one corresponding to  $-H \perp ab$ , which suggests that the  $B_z$  component solely should not result in the hysteresis. Meanwhile, if we only apply the in-plane field, Hall signal is not expected. So the interaction between Josephson vortices and pancake vortices is essential for the sign reversal of  $\rho_{xy}$  in the mixed state via rotation of the field. We would like to point out that the field corresponding to the maximum  $|\Delta\rho_{xx}|$  is just the field where the vortices move from the plastic to the elastic mode [31] as seen from the  $d\rho_{xx}/dB(B)$  curve in the inset of Fig. 2(b).

An alternative method to modify the pinning properties of the sample consists of doping the LCCO compound with Co atoms. In this case the Co substitution in place of Cu leads to a substantial reduction of the critical temperature with the consequent decrement of the critical current.

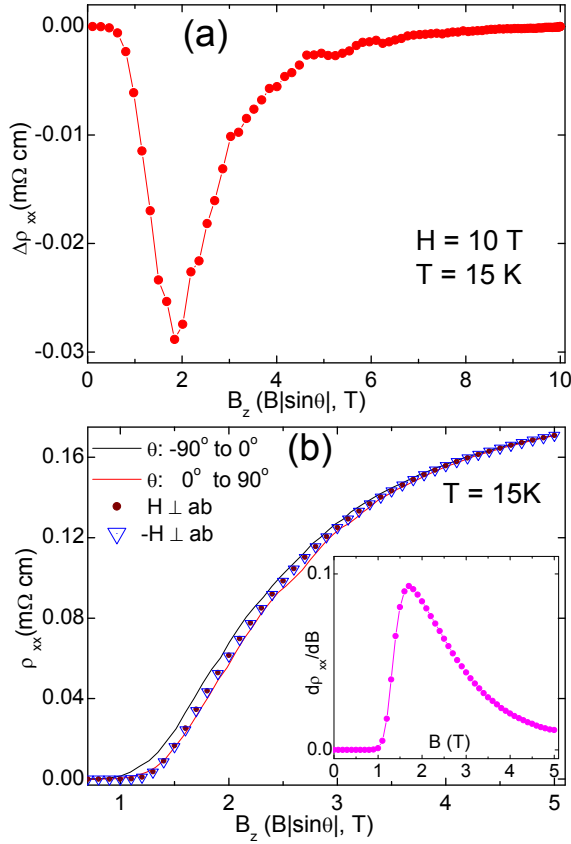


Fig. 2. (Color online) (a) The resistivity difference between the different field orientation, i.e.,  $\Delta\rho_{xx} = \rho_{xx,+\theta} - \rho_{xx,-\theta}$ , versus the field  $B_z$  at  $H=10$  T and  $T=15$  K. (b) The longitudinal resistivity  $\rho_{xx}$  versus the field  $B_z$  in LCCO films at  $T=15$  K by different changing methods. The insert in (b) shows  $d\rho_{xx}/dB$  versus the magnetic field  $B$  perpendicular to the  $ab$ -plane.

Fig. 3 shows isothermal curves  $\rho_{xx}(B)$  and  $\rho_{xy}(B)$  for LCCO:Co obtained with the applied magnetic field perpendicular to the  $ab$ -plane. Fig. 3(a) and (c) show both  $\rho_{xx}(B)$  and corresponding  $\rho_{xy}(B)$  curves at temperatures below  $T_{c0}$ , i.e., the critical superconducting transition temperature at zero field. In this temperature range, and for low enough fields vortices remain pinned thus leading to  $\rho_{xx}(B) = \rho_{xy}(B) = 0$ . As  $B$  increases,  $\rho_{xx}(B)$  follows a smooth increase towards the normal state resistivity value whereas  $\rho_{xy}(B)$  exhibits a more complex dependence with one ( $T > 6$  K) or two ( $T = 5$  K) sign reversals. It is important to mention that we do not observe double sign reversal in the mixed state of LCCO [18]. For temperatures higher than  $T_{c0}$  [see Fig. 3(b) and (d)],  $\rho_{xy}(B)$  exhibits a finite value at  $B = 0$ , as shown in Fig. 3(d). This is not surprising since the interaction between ferromagnetic

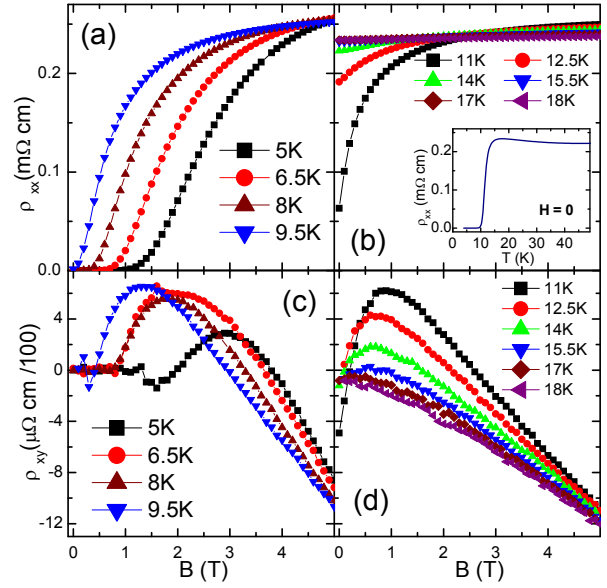


Fig. 3. (Color online) Longitudinal resistivity  $\rho_{xx}$  (a)-(b) and the Hall resistivity  $\rho_{xy}$  (c)-(d) versus the magnetic field perpendicular to the  $ab$ -plane of LCCO:Co thin films at different temperatures. Inset of (b):  $\rho_{xx}$  versus  $T$  at zero field.

Co atoms leads to a ferromagnetic state which in turn produces a finite internal magnetic field, even though the external magnetic field is zero[23,24].

As we have demonstrated in a previous report [23], the saturation magnetic field for LCCO:Co thin films is  $\sim 0.1$  T. This saturation field is much smaller than the values ( $\sim 1$  to 4 T) at which the sign reversal occurs, which suggests that there is no correlation between the intrinsic ferromagnetic state and the double sign reversal. Nevertheless, the Co ions should have preferentially the same magnetization polarity as the vortices in this field region. The doping by magnetic ions can significantly weaken the pinning strength in the LCCO thin films, which results in the clear transition of the sign reversal of the Hall resistivity  $\rho_{xy}$  from once to twice as we have discussed above.

In Fig. 4, we show the Hall resistivity  $\rho_{xy}$  versus  $B_z(B|\sin\theta|)$  in LCCO:Co thin films with different modes of varying the magnetic field. It can be seen that the minimum and maximum value with rotating field are shifted to  $\sim 1.9$  and 4.5 T, which are larger than those at 1.5 and 3 T by mode A with  $\mathbf{B} \perp ab$ -plane. Besides that, the value of  $\rho_{xy}(B_z)$  with rotating field is much larger than that by mode A with  $\mathbf{B} \perp ab$ -plane, which can be attributed to not only the coupling between the pancake vortices and the in-plane Josephson vortices, but also to the pres-

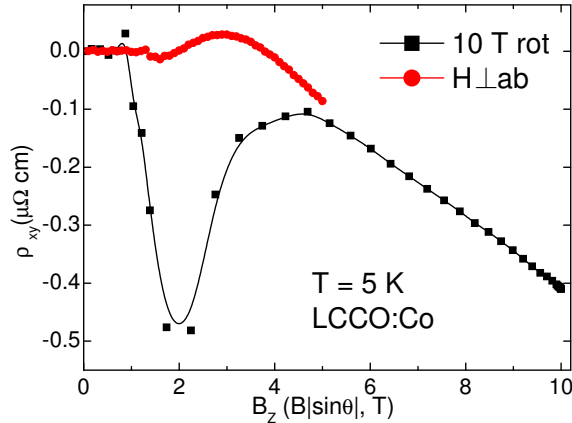


Fig. 4. (Color online) Hall resistivity  $\rho_{xy}$  versus  $B_z$  ( $B|\sin\theta|$ ) in LCCO:Co thin films at 5 K via different methods, i.e., by rotating the field orientation or changing the field strength but keeping  $B \perp ab$ -plane.

ence of the remanence magnetization in LCCO:Co samples during the rotation of the magnetic field.

#### 4. Conclusion

In summary, we have successfully introduced two different methods to tune the pinning strength in the samples and we do find significant changes in the Hall sign reversals from once to twice, which coincides with the change of the pinning environment from strong to weak. Our experimental results are in agreement with the prediction of the theoretical and numerical works [14,20]. The double sign reversal in the Hall resistivity has been revealed in the electron-doped LCCO thin films via two different modes in the present investigation. In the case of substituting Co for Cu in LCCO thin films, the appearance of the double sign reversal of the Hall resistivity is consistent with a weaker pinning scenario. While, by tilting the magnetic field in the LCCO thin films, the double sign reversal of  $\rho_{xy}(B)$  is caused by the coupling between the pancake vortices and in-plane Josephson vortices.

#### 5. Acknowledgments

We acknowledge the support from the MOST 973 projects No. 2011CBA00110 and 2009CB930803, and the National Natural Science Foundation of China and the bilateral China/Flanders Project. This work was also supported by Methusalem Funding by the Flemish government, FWO-Vlaanderen, and the Belgian Inter-University Attraction Poles

IAP Programmes. A.V.S. is grateful for the support from the FWO-Vlaanderen.

#### References

- [\*] email address: beiyi.zhu@iphy.ac.cn
- [1] Y.S. Oh, K. H. Kim, P.A. Sharma, N. Harrison, H. Amitsuka, and J.A. Mydosh, *Phys. Rev. Lett.* **98**, 016401 (2007).
- [2] John Singleton, *"Band-theory and electronic properties of solids"*, Oxford University Press, New York, USA (2001).
- [3] F.F. Balakirev, J.B. Betts, A. Migliori, S. Ono, Y. Ando, and G.S. Boebinger, *Nature* **424**, 912 (2003).
- [4] D. LeBoeuf, N.D. Leyraud, J. Levallois, R. Daou, J.-B. Bonnemaison, N.E. Hussey, L. Balicas, B.J. Ramshaw, R.X. Liang, D. A. Bonn, W.N. Hardy, S. Adachi, C. Proust, and L. Taillefer, *Nature* **450**, 533 (2007).
- [5] C. Pfeleiderer and R. Hackl, *Nature* **450**, 490 (2007).
- [6] V.M. Vinokur, V.B. Geshkenbein, M.V. Feigel'man and G. Blatter, *Phys. Rev. Lett.* **71**, 1242 (1993).
- [7] W.N. Kang, D.H. Kim, S.Y. Shim, J.H. Park, T.S. Hahn, S.S. Choi, W.C. Lee, J.D. Hettinger, K.E. Gray, and B. Glagola, *Phys. Rev. Lett.* **76**, 2993 (1996).
- [8] W. Göb, W. Liebich, W. Lang, I. Puica, R. Sobolewski, R. Rössler, J. D. Pedarnig, and D. Bäuerle, *Phys. Rev. B* **62**, 9780 (2000).
- [9] Y. Matsuda, T. Nagaoka, G. Suzuki, K. Kumagai, M. Suzuki, M. Machida, M. Sera, M. Hiroi, and N. Kobayashi, *Phys. Rev. B* **52**, R15749 (1995).
- [10] S.J. Hagen, C.J. Lobb, R.L. Greene, and M. Eddy, *Phys. Rev. B* **43**, 6246 (1991).
- [11] W.N. Kang, B.W. Kang, Q.Y. Chen, J.Z. Wu, Y. Bai, W.K. Chu, D.K. Christen, R. Kerchner, and S.I. Lee, *Phys. Rev. B* **61**, 722 (2000).
- [12] J.E. Hirsch and F. Marsiglio, *Phys. Rev. B* **43**, 424 (1991).
- [13] A.G. Aronov and S. Hikami, *Phys. Rev. B* **41**, 9548 (1990).
- [14] Z.D. Wang, J.M. Dong, and C.S. Ting, *Phys. Rev. Lett.* **72**, 3875 (1994).
- [15] B.Y. Zhu, D.Y. Xing, Z.D. Wang, B.R. Zhao, and Z.X. Zhao, *Phys. Rev. B* **60**, 3080 (1999); B.Y. Zhu, *Physica C* **276**, 309 (1997).
- [16] R. Wordenweber, E. Hollmann, J. Schubert, R. Kutzner, Ajay Kumar Ghosh, *Appl. Phys. Lett.* **94**, 202501 (2009).
- [17] N.P. Armitage, F. Ronning, D.H. Lu, C. Kim, A. Damascelli, K.M. Shen, D.L. Feng, H. Eisaki, Z.X. Shen, P.K. Mang, N. Kaneko, M. Greven, Y. Onose, Y. Taguchi, and Y. Tokura, *Phys. Rev. Lett.* **88**, 257001 (2002).
- [18] K. Jin, B.Y. Zhu, J. Yuan, H. Wu, L. Zhao, B.X. Wu, Y. Han, B. Xu, L.X. Cao, X.G. Qiu, and B.R. Zhao, *Phys. Rev. B* **75**, 214501 (2007).
- [19] Z.D. Wang and C.S. Ting, *Phys. Rev. Lett.* **67**, 3618 (1991); *Phys. Rev. B* **46**, 284 (1992).
- [20] B.Y. Zhu, *Mod. Phys. Lett. B* **10**, 1227 (1996).
- [21] B.X. Wu, K. Jin, J. Yuan, H.B. Wang, T. Hatano, B.R. Zhao, and B.Y. Zhu, *Supercond. Sci. Technol.* **22**, 085004 (2009).

- [22] B.X. Wu, K. Jin, J. Yuan, H.B. Wang, T. Hatano, B.R. Zhao, and B.Y. Zhu, *Physica C* **469**, 1945 (2009).
- [23] K. Jin, J. Yuan, L. Zhao, H. Wu, X.Y. Qi, B.Y. Zhu, L.X. Cao, X.G. Qiu, B. Xu, X.F. Duan, and B. R. Zhao, *Phys. Rev. B* **74**, 094518 (2006).
- [24] K. Jin, L. Zhao, H. Wu, J. Yuan, S. J. Zhu, L. J. Gao, B. Y. Zhu, B. Xu, L. X. Cao, X. G. Qiu, B. R. Zhao, *Physica C* **460**, 410 (2007).
- [25] K. Jin, B.Y. Zhu, B.X. Wu, L.J. Gao, and B. R. Zhao, *Phys. Rev. B* **78**, 174521 (2008).
- [26] K. Jin, N. P. Butch, K. Kirshenbaum, J. Paglione and R. L. Greene, *Nature* **476**, 73 (2011).
- [27] A.N. Grigorenko, S.J. Bending, I.V. Grigorieva, A.E. Koshelev, T. Tamegai, and S. Ooi, *Phys. Rev. Lett.* **94**, 067001 (2005).
- [28] A.E. Koshelev, *Phys. Rev. B* **71**, 174507 (2005).
- [29] T. Matsuda, O. Kamimura, H. Kasai, K. Harada, T. Yoshida, T. Akashi, A. Tonomura, Y. Nakayama, J. Shimoyama, K. Kishio, T. Hanaguri, and K. Kitazawa, *Science* **294**, 2136 (2001).
- [30] M. Amirfeiz, M.R. Cimberle, C. Ferdeghini, E. Giannini, G. Grassano, D. MarrY, M. Putti, A.S. Siri, *Physica C* **288** 37 (1997).
- [31] N.-C. Yeh, *Phys. Rev. B* **43**, 523(1991).